

## Integrated Mesoscale Architectures for Sustainable Catalysis (IMASC)

EFRC Director: Cynthia Friend

Lead Institution: Harvard University

Start Date: August 2014

**Mission Statement:** *To drive and conduct transformative research in mesoscale science for sustainable catalysis, with full integration of multi-scale experimental, theoretical and computational approaches.*

IMASC is seeking fundamental understanding for the purpose of developing novel mesoporous catalysts (non-zeolite) for sustainable processes that drive the conversion and production of platform chemicals. Understanding reaction kinetics, building innovative catalyst architectures and improving reaction selectivity under catalytic conditions is leading to new opportunities to decrease fuel consumption through creation of more efficient processes. IMASC is advancing the scientific grand challenge question “how do we design and perfect atom- and energy-efficient synthesis of revolutionary new forms of matter with tailored properties?” through integrated efforts at Harvard University, Tufts University, Lawrence Berkeley and Lawrence Livermore National Laboratories, and the Fritz Haber Institute in Germany.

### IMASC Strategic and Research Objectives:

#### IMASC Strategic Objectives:

- To develop principles for designing efficient catalytic processes based on fundamental understanding from experiment and theory
- To apply, test and refine these principles on mesoporous materials under working catalytic conditions.

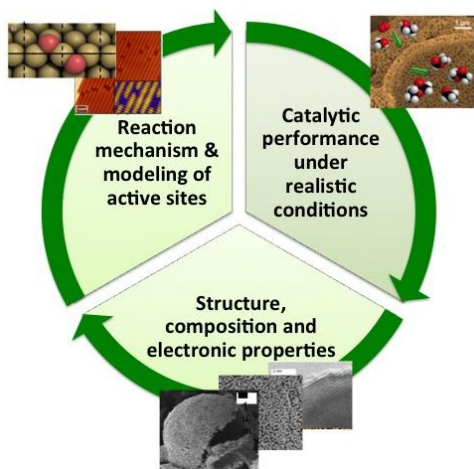


Fig. 1: A schematic representation of IMASC's approach to develop mesoscale architectures for sustainable catalysis

IMASC research is strategically organized into three Focus Areas (Fig. 1) capitalizing on a judiciously chosen combination of technical skill sets that are designed to address the following **research objectives** necessary to achieve our strategic objectives, mission and vision:

1. Delineate reaction mechanisms of selective oxidation, hydrogenation and dehydrogenation reactions in order to establish paradigms for predicting selectivity and activity.
2. Understand the function of dilute alloy surfaces and develop models for predicting materials compositions to optimize activity and selectivity.
3. Advance the methodology for studies under reaction conditions—vapor and liquid phase—in order to relate catalytic function to surface composition, structure and electronic properties.
4. Identify the design elements in building mesoscale catalyst architectures and establish principles for their controlled synthesis.
5. Parameterize structure-activity-selectivity relationships to develop models for rapidly selecting catalyst materials and reaction conditions for specific reactions.

### IMASC Research Team and Research Focus Areas:

The IMASC team includes chemists, physicists, chemical engineers, and materials scientists who are performing research organized into three research Focus Areas: (1) Reaction Mechanisms and Modeling of Active Sites; (2) Understanding Performance under Catalytic Conditions; and, (3) Structure, Composition and Electronic Properties (cross-cutting theme).

**Focus Area 1 (FA1): Reaction Mechanisms and Modeling of Active Sites.** The mechanisms of selective catalytic hydrogenation and oxidation are being investigated on the molecular scale on well-defined single crystal surfaces and on complex mesoporous materials under highly controlled conditions (Figure 1). Studies are leading to a comprehensive model of reaction mechanisms, including elementary chemical steps and the nature of active sites. Investigations underway include a range of (host) materials, including Cu, Ag, and Au combined with active metals (e.g., Pd, Ni and Pt). Designed meso- and nanoporous materials are synthesized as catalysts. FA1 is focusing on relating reaction selectivity to materials composition and structure as we continue to build up a sophisticated general model for these classes of reactions. Atomistic theory is providing insight into the interplay of electronic and geometric structure in determining bonding and reactivity of molecular intermediates. This research is beginning to inform catalyst design through close coupling with research in the other Focus Areas.

**Focus Area 2 (FA2): Understanding Performance under Catalytic Conditions.** The efficacy of specific mesoporous architectures for catalytic activity and selectivity under realistic reaction conditions is being established through reactor studies. The unique capability of temporal analysis of products (TAP) experiments to quantitatively determine rate constants is combined with studies in flow reactors for vapor phase processes *in situ* (1 atm. pressure). This research is closely coupled with the research in FA1 to refine the working hypotheses about what materials properties and reaction conditions yield better performance (Fig. 1). Close coupling with FA 3 is used to model the effect of mesoporous geometry to the local surface concentrations of reactants and products through multi-scale simulations.

**Focus Area 3 (FA3): Understanding Structure, Composition and Electronic Properties under catalytic conditions.** This cross-cutting research theme is enabling development and use of advanced theoretical and experimental tools for understanding the effects of surface composition, structure and electronic properties on the catalytic reactivity of the model and mesostructured materials and the scale-up from the atomic scale to the mesoscale architectures. Powerful microscopies and x-ray probes are combined to understand the working reactive surfaces. Multiscale theory is providing a fundamental understanding of both the materials synthesis and the evolution of the mesoscale materials under reactive conditions.

Integrated Mesoscale Architectures for Sustainable Catalysis (IMASC)	
Harvard University	Cynthia Friend (Director), Efthimios Kaxiras (Deputy Director), Challa Kumar (Managing Director), David C. Bell, Robert Madix
Tufts University	Maria Flytzani-Stephanopoulos, E. Charlie H. Sykes
Lawrence Berkeley National Laboratory	Miquel Salmeron
Lawrence Livermore National Laboratory	Juergen Biener

**Contact:** Cynthia Friend, Director, [friend@fas.harvard.edu](mailto:friend@fas.harvard.edu)  
617-495-4052, <http://efrc.harvard.edu>